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Disintegration of $_{56}Ba^{131} \rightarrow {}_{55}Cs^{131} \rightarrow {}_{54}Xe^{131}$

By Evani Kondaiah

With 6 figures in the text

1. Introduction

Katcoff (1) in 1947 reported the formation of Ba–Cs chain of probable mass assignment 131 by neutron activation of Ba. He also reported that Ba^{131} decays predominantly by orbital electron capture with a half-life of 12 days emitting γ-radiation of about 0.26 MeV, 0.5 MeV and roughly 1.2 MeV (low intensity) and that Cs^{131} decays exclusively by K-electron capture with a half-life of 10.2 days emitting no detectable γ-rays.

In the same year Fu-Chen Yu (2) et al. reported 220 ± 10 keV, 500 ± 15 keV and 1.7 ± 0.1 MeV γ-rays in Ba^{131} and also that Cs^{131} emits a highly converted γ-ray of energy 145 ± 10 keV; conversion in this case was reported as 97 %.

Recently L. Yaffe (3) et al. confirmed the result of Fu-Chun Yu et al. viz. that Cs^{131} emits a highly converted γ-ray of the above mentioned energy.

Finkle (4) found that neither Ba^{131} nor Cs^{131} emits positrons.

Recently, at the conference of the Swedish National Committee for Physics, I reported on the electron spectrum of Cs^{131}, showing several groups of electrons. More recent investigations, however, have revealed that these lines in fact belong to Ba^{131}.

2. Preparation of samples

Barium nitrate was irradiated in the Harwell pile* and the chemical separations were carried out in the Radio-Chemical Laboratory of this institute by Fil. mag. W. Forsling as follows:

The irradiated barium nitrate was dissolved in water and about 7 mg of rubidium chloride was added. A slight excess of ammonia was added to the solution and barium carbonate was precipitated by am. carbonate. The barium carbonate was dissolved in dilute hydro-chloric acid and the precipitation was repeated. The filtrates were evaporated to dryness and the ammonium salts were expelled by cautious ignition. The residue was then dissolved in water and a few drops of ammonia, am. carbonate and am. oxalate were added to remove the remaining Ba. After some hours the barium carbonate was removed as before.

In order to get Cs. quite free from any Ba-activity, it was found necessary to add inactive Ba-carrier to the filtrate twice and each time barium carbonate was pre-

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* I wish to express my thanks to Dr. Seligman, for this irradiation.

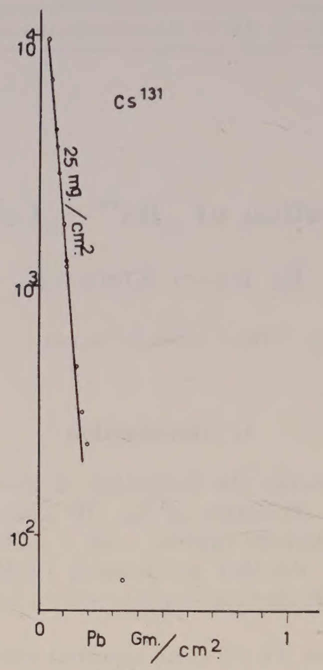

Fig. 1.

cipitated as mentioned above. The alkali-chlorides were dissolved in water and 1 mg of caesium chloride was added. The Cs was separated from Rb. by the antimony-tri-chloride-ferric chloride method (5). The filtrate which seemed to contain most of the Cs, was then evaporated to dryness. Caesium chloride powder thus got was dissolved in a few drops of water and evaporated on to the sample holder.

Part of the above reprecipitated barium carbonate was dissolved in dilute hydrochloric acid and the solution was evaporated to dryness. Barium chloride was dissolved in water to which a few drops of acetic acid and solution of am. acetate were added. Strontium chloride and calcium chloride were then added. The precipitate of barium chromate thus got was allowed to settle down and filtered off and washed with dilute am. chromate solution and water. The precipitate was rinsed into a beaker with warm nitric acid which dissolved the chromate. The solution was then diluted and boiled. Fresh amounts of Sr. and Ca-carriers were added. The solution was then neutralised with am. acetate and the barium chromate was precipitated and washed as before. The chromate was reduced in hydro-chloric acid. The solution was boiled and ammonia was added in a slight excess before precipitating with am. sulphide. The precipitate was filtered off and washed. Then the filtrate was boiled, made slightly acidic with hydro-chloric acid for removing the hydrogen sulphide. The Ba. was then precipitated as carbonate which was dissolved in dilute hydrochloric acid. The acid was evaporated and the barium chloride thus got, was transferred over to the sample holder.

The sample holder consists of an ebonite ring of 1.8 cm diameter. Glasan paper of 6 mg/cm^2 thick was fastened to this ring by zapon-lac solution. Except in the case of studies made in the low energy region, generally the sample was made into a fine

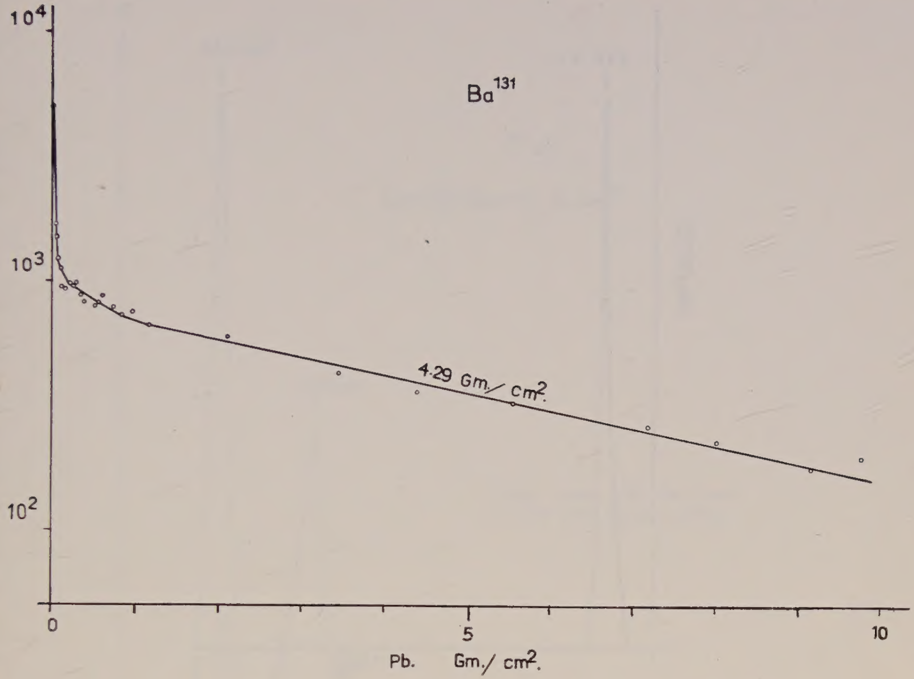

Fig. 2.

powder and was fixed on the glasan paper using one or two drops of very dilute zapon-lac solution. In the case of samples studied in the low energy region the caesium-chloride powder was dissolved in a few drops of water, which was then evaporated on to the sample holder using infra red light.

3. Half-life and absorption measurements

The Ba^{131} and Cs^{131} decays were followed for two half-life periods in each case. They come out as 13 and 10 days respectively. These are in fundamental agreement with the previously reported values.

Fig. 1 gives the absorption curve in Pb for Cs^{131} radiations. Fig. 2 gives the same for Ba^{131} radiations. The straight line, giving a half-thickness of 25 mg/cm², in the case of Cs. corresponds to X-rays. The curve for Ba consists of more than one component, the hardest component corresponding to a half-thickness of 4.29 gm/cm² i.e. to 0.49 MeV γ-rays. The softer portion corresponds to the other soft γ-rays and X-rays. Thus it seems there are no unconverted γ-rays emitted by the Cs. isotope and also no γ-rays of higher energy than 0.49 MeV are emitted by Ba. isotope.

4. Electron spectra

Figs. 3 and 4 are the electron spectra of Cs^{131} and Ba^{131}, respectively. The resolution of the spectrometer (6) was set to be 4.5 %. The inner spectrum in Fig. 4 was taken with the same Ba. sample in the same spectrometer under similar conditions

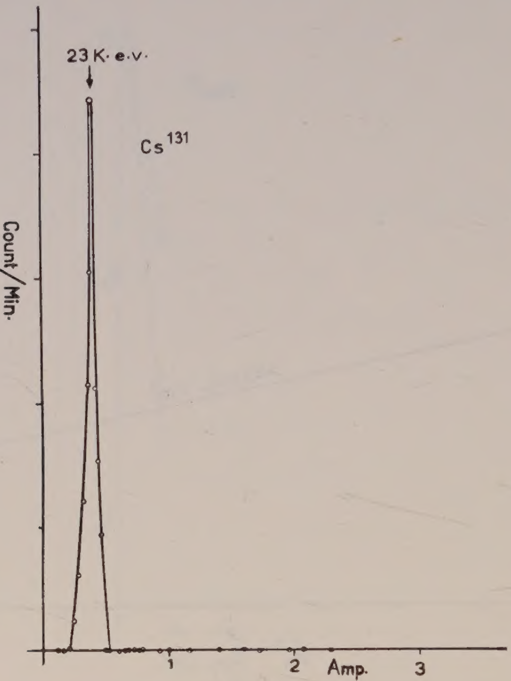

Fig. 3.

after 16 days decay after the outer one had been taken. By comparing the areas one can see that all the γ-rays are decaying with the same half-life. In fig. 4, the number of counts per minute was divided by the current and the result N, is used as ordinate, whereas in fig. 3, counts per minute, itself is used as ordinate. Fig. 4 shows four γ-rays more or less converted and the energies of the corresponding electron peaks are given viz. 84.5, 116.6, 172.7, 334.0 and 456.0 KeV. The first two are the K and L lines of 121 keV γ-ray and the rest are the K-lines of 209, 370 and 492 keV γ-rays respectively. The line at 23.4 keV is an Auger electron line corresponding to the (K–L–L′) energy. The electron spectrum of Cs^{131} (fig. 3) consists of one group at 23 keV, which is again an Auger electron line. Thus, the Cs^{131} isotope emits no converted γ-rays. These electron spectra were followed several times in two spectrometers, one of which has got high intensity (7) at about the same resolution. The G. M.-counter window was made of nylon and could transmit electrons down to about 10 keV. Cs^{131} was specially investigated to see if there were any positrons of low energy. No positive results have been obtained in this connection. However the possibility that the Auger electrons may be masking any soft positron emission cannot be excluded. The same holds for Ba^{131} isotope as well.

5. γ-spectrum

The method followed in this case is the usual one. The Ba-sample was put in a copper capsule with a copper foil of 0.1 mm thick to absorb the highest energy internal conversion electrons (456 keV). Over this copper foil a thin Pb-foil was placed

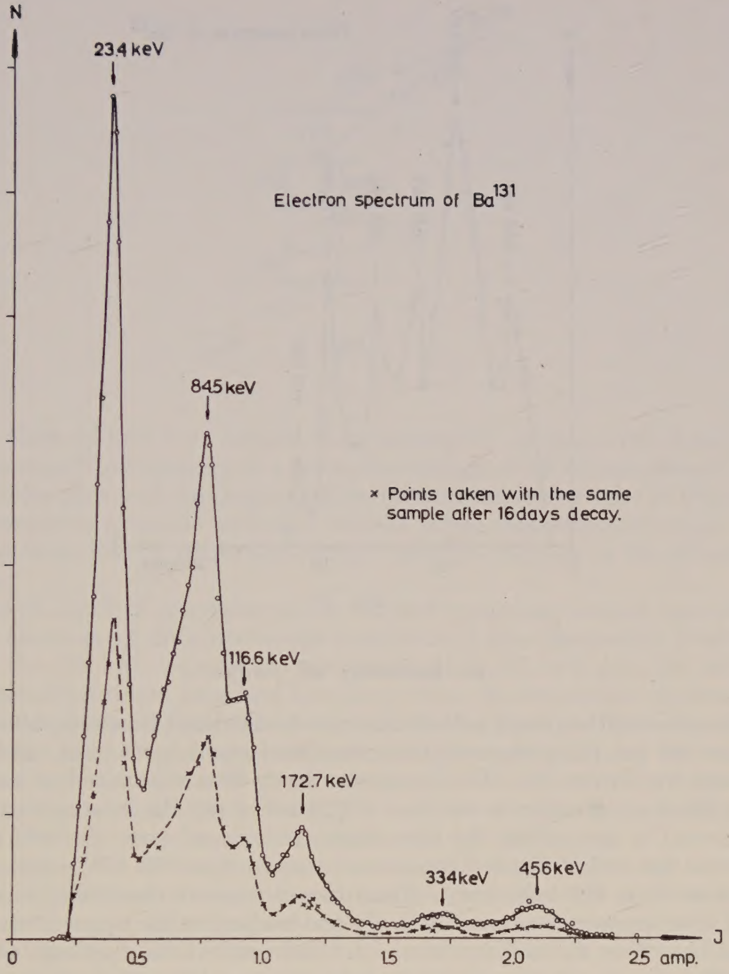

Fig. 4.

for the photo effect to take place. Lead foils of 0.03 and 0.01 mm thick were used. Lead foils of 0.03 mm thick were used to see whether there are any unconverted γ-rays of high energy (1.7 MeV^2 or so). Fig. 5 gives the photo spectrum taken with the 0.03 mm thick lead foil. The photo-electron groups at 34.8, 116, 188, 285, 411, and 483 keV respectively are the K-line of 122 keV, K-line of 204 keV, L-line of 204 keV, K-line of 373 keV, K-line of 499 keV, L-line of 499 keV γ-rays. The L-lines of 122 keV and 373 keV γ-rays are overshadowed by the K-lines of 204 keV and 499 keV γ-rays, respectively. The broadening of the K-line at 116 keV is due to the contribution of the L-line of 122 keV γ-ray and to the thickness of the radiator used.

Thus, the photo-electron groups observed give the same values for γ-ray energies within the experimental error, as derived from the internal conversion electron groups. The energies of the γ-rays are therefore fairly well established. These are 122 ± 2 keV, 206 ± 4 keV, 372 ± 3 keV, 494 ± 6 keV.

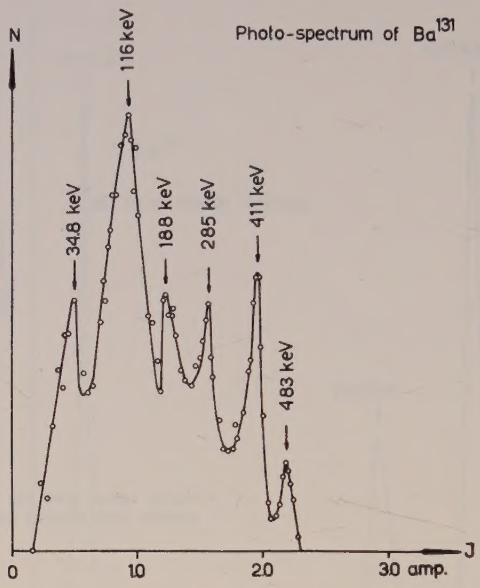

Fig. 5.

6. Intensity of γ-rays

The intensities of the γ-rays in Ba^{131} have been calculated from the photo-spectrum. The values for the photo-electric absorption coefficient have been taken from the graph given by Evans (8). The intensities have been calculated as suggested by Deutsch (9) et al. Except in the case of 122 keV γ-ray the internal conversion has been neglected in computing the intensities. It is found that 494 keV γ-ray is the most intense one and is about 5 times more intense than 206 keV γ-ray and 4 times more intense than 372 keV γ-ray. From this it appears that there is a branching in orbital electron capture decay of Ba^{131}, one leading to the upper level of 206 keV γ-ray and the other to its lower level. A third one, which may lead to the upper level of 122 keV γ-ray is thought to be improbable as 122 keV γ-ray is not found to be more intense than any of the other γ-rays, and furthermore the γ–γ-coincidence rate calculated according to the intensities estimated here has tallied well with the experimentally observed value. The percentages given in the decay scheme (fig. 6) correspond to the relative intensities. However, these must be taken as estimates only.

7. Coincidence measurements

(a) e^-, e^- coincidences

Coincidences between the internal conversion electrons of the different γ-rays have been investigated in the spectrometer (7). For this purpose the sample holder chamber was demounted and a chamber holding a β-tube was substituted in its place. The sample was supported just in front of this β-tube and Al-foils of different thicknesses were interposed in between the sample and the β-tube. It was possible to

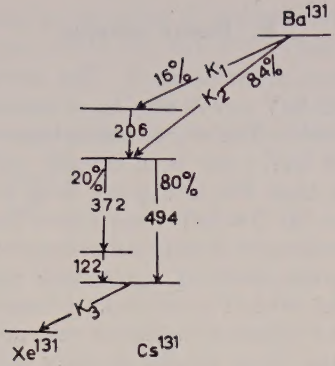

Fig. 6.

manipulate these Al-foils from outside the spectrometer, so that a foil of any required thickness to cut off electrons up to a particular energy could be introduced with ease. Thus one of the groups of electrons could be focused into the β-tube at the other end of the spectrometer using the necessary current in the magnetic coils, while electrons of energy, greater than a particular energy could be counted in the β-tube behind the sample.

When the 84.5 keV K-electrons of the 122 keV γ-ray are focused into the far off β-tube and electrons of different γ-rays were allowed into the nearby β-tube, it was found that the 122 keV γ-ray is in coincidence with 372 keV and 206 keV γ-rays, while the coincidence rate remained the same when the conversion electrons of 494 keV γ-ray were allowed into or cut-off from the nearby β-tube. When the conversion electrons of the 372 keV γ-ray were allowed into the nearby β-tube, the coincidence rate increased and when 206 keV γ-ray was also allowed, the coincidence rate almost doubled showing that both 206 keV and 372 keV γ-rays are in cascade with 122 keV γ-ray while 494 keV γ-ray is not in cascade with 122 keV γ-ray.

However, when the internal conversion electrons of 206 keV γ-ray were focused into the far off β-tube and coincidences examined between this and 494 keV γ-ray conversion electrons, no coincidence rate higher than the back-ground could be found. This is probably due to the conversion in the case of 206 keV and 494 keV γ-rays being very small. Whatever few coincidences that could have been found, might have been masked by the back-ground coincidences between X-rays and the conversion electrons of 206 keV γ-ray. The resolving time of the coincidence circuit used was 0.4×10^{-6} sec.

(b) γ–γ-coincidences

The same coincidence circuit with the same resolving time was used in conjunction with two calibrated γ-tubes in a standard set up. These γ-tubes have gold cathodes of 0.1 mm thick. These tubes are mounted side by side in the centre of which the sample is mounted. The back-ground coincidence rate due to cosmic rays as well as the chance coincidence rate, have been taken into account. X-rays (32 keV) cannot penetrate these γ-tubes with any appreciable intensity. The number of γ–γ-coincidences per recorded γ is found to be $(0.23 \pm 0.02) \times 10^{-3}$. The γ–γ-coincidence effect to be expected from the proposed decay scheme (Fig. 6) with the relative intensities mentioned there, comes out as 0.205×10^{-3}, i.e. in good agreement with the observed value.

8. Decay scheme

The decay scheme has been given in fig. 6. The energy measurements of γ-rays suggest that 372 keV and 122 keV γ-rays may be in cascade with a cross-over transition γ-ray of energy 494 keV. The e^-, e^--coincidence experiments confirm this. Further they show that 206 keV γ-ray is in cascade with 122 keV γ-ray. The intensity measurements show that 206 keV γ-ray is of less intensity than 494 keV γ-ray. These two facts place the 206 keV γ-ray above the common level of 372 keV and 494 keV γ-rays. A branching of K-capture is expected from the intensity measurements. γ–γ-coincidence rate observed, agrees well with the value calculated according to this picture. If the 494 keV γ-ray is placed separate from the other γ-rays, the γ–γ-coincidence rate to be expected comes out very much lower than the observed value. So, it appears that the decay scheme is fairly well established taking all the observed facts into account. Cs^{131} decays exclusively by orbital electron capture and emits no γ-rays. It has been shown as such in fig. 6. The percentages given in the figure refer to the relative intensities.

9. Internal conversion

It is possible, in this case, to estimate the conversion of each of the γ-rays, since the K to L conversion ratio at least in one case (122 keV γ-ray) can be known from the conversion electron spectrum of Ba^{131} (Fig. 4). The K to L conversion ratio in the case of 122 keV γ-ray comes out to be about 3.5. HEBB and NELSON's (10) curves should be lowered for 'Z' higher than 35. So, $1 = 2$ curve fits for the above ratio. The K to L conversion ratios, on the assumption that the γ-radiation (122 keV), is electric quadrapole or magnetic dipole, have been calculated from theory. They come out as 4.7 if electric 2^2 pole, 5.5 if magnetic 2^2 pole and 6.3 if magnetic 2^1 pole. So, it appears that 122 keV γ-ray consists exclusively of electric 2^2 pole radiations. On this assumption the internal conversion coefficient comes out to be 80 % using Rose's tables.*

Using this value it is possible to estimate the total number of 122 keV γ-rays emitted by the sample. From it, using the relative intensities of γ-rays given in Fig. 6., it is possible to estimate the total number of γ-rays of each energy. The number of converted γ-rays of each energy can be known from the conversion electron spectrum (Fig. 4). From these, the conversion coefficient in each case can be estimated. They come out as 15 %, 3 % and 1 % for 206, 372 and 494 keV γ-rays respectively.

10. Multipole order of γ-rays

The K-shell internal conversion coefficients of the γ-rays have been calculated from Rose's tables*, assuming different values for l. These values for electric 2^2 pole radiations (α_2) and magnetic 2^1 pole radiations (β_1) have been given in Table 1. The total internal conversion coefficients estimated from experiment have been given in the last column of the table. If L-conversion is also taken into account, the theoretical values α_2 and β_1 may agree much better with the experimentally estimated values. Even now, the agreement between theoretical values and experimental values is quite good as L-conversion for all the γ-rays except 122 keV γ-ray is negligible.

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* "Tables of K-shell internal coefficients" by M. E. ROSE, G. GOERZEL, B. SPINRAD, J. HARR, P. STRONG. Private communication.

Thus all these γ-rays appear to be either 2^2 pole electric or 2^1 pole magnetic or a combination of these two types while 122 keV γ-rays may be of exclusively electric quadrapole radiations.

Table 1

Energy	$K = \frac{\text{Energy}}{mc^2}$	α_2	β_1	Experimental Total internal conversion coefficient
122 keV	0.24	0.64	0.425	0.8
206 »	0.40	0.12	0.106	0.15
372 »	0.73	0.019	0.0220	0.03
494 »	0.97	0.0095	0.0107	0.01

11. Discussion

It is interesting to note that this chain involves two consecutive orbital electron captures without emitting positrons. As mentioned elsewhere, the only possibility of any low energy positrons of small intensities is still open. However, as no annihilation radiation (0.51 MeV) γ-rays are found either in Ba^{131} or in Cs^{131} samples it can reasonably be assumed that there are no positrons in either case.

It is not inconsistent with the disintegration scheme proposed here to assume another K-capture transition from Ba^{131} level to the ground level of Cs^{131}. It is not shown in fig. 6 as there has been no necessity of assuming such a transition.

In the case of Cs^{131} decay, the result obtained here is in agreement with that of KATCOFF (1) and is contradicting the results of FU-CHEN YU (2) and YAFFE (3). Probably these latter investigators might have got Ba. impurities in their Cs.-samples. No γ-ray of higher energy than 494 keV is observed in the case of Ba^{131}. It may be mentioned for clarity that the absorption curve of Ba^{131} given in Fig. 2 has, in fact, been followed till the count has dropped to the back-ground count; this has occurred after using 20 gm/cm² of lead. Thus, neither the absorption curve in lead (Fig. 2), nor the electron spectrum (Fig. 4), nor the photo-spectrum (Fig. 5) indicate such a high energy γ-ray as 1.2 MeV reported by KATCOFF (1) or 1.7 MeV reported by FU-CHEN YU (2). This may be due to the comparatively low specific activity of the samples I used. However, from my measurements it may be estimated that if at all such a γ-ray of high energy exists, its intensity should be $\lesssim 5\,\%$ that of the 494 keV γ-ray.

12. Summary

1. The energies of γ-rays in Ba^{131} have been determined from electron spectrum and photo-spectrum taken in a spectrometer. They are 122 ± 2 keV, 206 ± 4 keV, 372 ± 3 keV, 494 ± 6 keV.

2. Intensities of γ-rays and branching ratio in K-capture have been estimated from the photo-spectrum of Ba^{131}.

3. e^-, e^--coincidences have been investigated in a spectrograph.

4. γ–γ-coincidence rate has been found to be $(0.23 \pm 0.02) \times 10^{-3}$ per recorded γ.

5. A probable disintegration scheme taking all the observed facts into consideration has been discussed.

6. Internal conversion coefficients and multipole order nature of γ-rays have been estimated.

Acknowledgements

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